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Title:	New Opportunity for Improved Nuclear Forensics, Radiochemical Diagnostics, and Nuclear Astrophysics: Need for a Total-Cross-Section Apparatus at the LANSCE
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New Opportunity for Improved Nuclear Forensics, Radiochemical Diagnostics, and Nuclear Astrophysics: Need for a Total-Cross-Section Apparatus at the LANSCE

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Total-cross-section measurements are feasible on a much wider range of radioactive samples than (n,γ) cross-section measurements, and information extracted from the former can be used to set tight constraints on the latter. There are many (n,γ) cross sections of great interest to radiochemical diagnostics, nuclear forensics, and nuclear astrophysics which are beyond the reach of current direct measurement, that could be obtained in this way. Our simulations indicate that measurements can be made at the Manuel Lujan Jr. Neutron Scattering Center at the Los Alamos Neutron Science Center for samples as small as 10 μg. There are at least 40 high-interest nuclides which should be measurable, including ^{88}Y, ^{167,168,170,171}Tm, ^{173,174}Lu, and ^{189,190,192}Ir.

There are many radionuclides for which (n,γ) cross sections are of great interest to radiochemical diagnostics, nuclear forensics, and nuclear astrophysics which are beyond the reach of current, or foreseeable future, direct measurement techniques. The main problem with direct measurements is that background from decay of the radioactive sample overwhelms the signal from (n,γ) events. In contrast, neutron-total-cross-section (transmission) measurements currently are feasible for many of these nuclides. In addition, such data provide information needed to accurately calculate (n,γ) cross sections via the nuclear statistical model (NSM).

With the very high flux at the Manuel Lujan Jr. Neutron Scattering Center (MLNSC) at the Los Alamos Neutron Science Center (LANSCE), such measurements should be feasible for samples as small as 10 μg. Samples of this size should be attainable for many nuclides of interest. Producing the radionuclides of interest will likely be the main challenge. On the other hand, fabricating, handling, and transporting the samples should be much less difficult than is the case for (n,γ) measurements. Furthermore, an initial survey indicates that several of the radionuclides of interest can be produced at the Isotope Production Facility at LANSCE.

The results in Fig. 1 illustrate that ^{151}Sm resonance parameters obtained from analysis of a transmission measurement made in 1975 [1] in the energy range below 300 eV can be used to obtain the capture cross section to an accuracy on par with direct measurements [2–4] made 30 years later, to energies as high as 300 keV.

According to the NSM, the average neutron capture cross section due to s waves can be written as,

$$\langle\sigma_\gamma\rangle = \frac{2\pi^2}{k^2}\frac{S_0\langle\Gamma_\gamma\rangle/D_0}{S_0+\langle\Gamma_\gamma\rangle/D_0}W, \tag{1}$$

where k is the wave number and W the width-fluctuation correction factor. All three parameters (neutron strength function S_0, average resonance spacing D_0, and average total radiation width $\langle\Gamma_\gamma\rangle$) needed for Eq. 1 were determined from $\mathcal{R}$-matrix analysis of transmission data taken with a 204.1-mg sample of ^{151}Sm, which resulted in resonance energies E_0 and reduced neutron widths $g\Gamma_n^0$ for 120 resonances below 300 eV, and total radiation widths

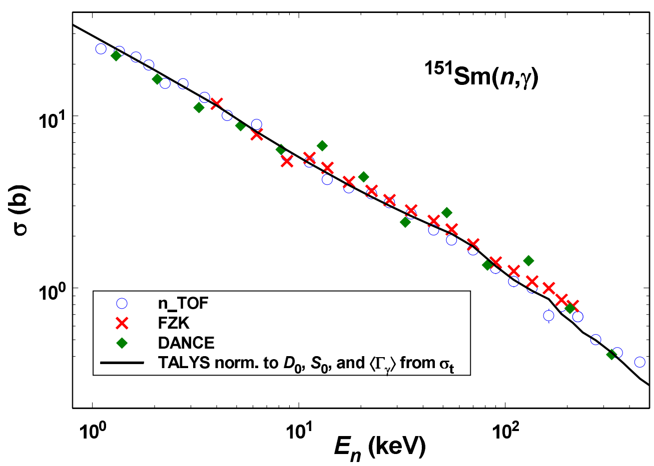

FIG. 1: ^{151}Sm(n,γ) cross section in the unresolved-resonance range. Symbols depict results from three different measurements [2–4], and the solid black curve is the cross section calculated by TALYS after adjustment to the average resonance parameters determined from an earlier ^{151}Sm+n total-cross-section measurement [1].

Γ_γ for 13 of the 14 lowest-energy resonances. These resonance parameters were used to obtain $S_0 = 3.58 \pm 0.59$, $D_0 = 1.1 \pm 0.07$ eV, and $\langle\Gamma_\gamma\rangle = 96.5 \pm 1.4$ meV in the usual manner [5], after correction for missed resonances [6]. The NSM code TALYS [7] was used to calculate the ^{151}Sm(n,γ) cross section after adjustment to these average resonance parameters. As can be seen in Fig. 1, the resultant NSM cross section is in excellent agreement with the available ^{151}Sm(n,γ) data [2–4] *across the entire measured energy range.*

The same approach can be used for other nuclides provided that sufficient (≈50) resonances are observed to obtain S_0, D_0, and $\langle\Gamma_\gamma\rangle$.

A transmission experiment is among the simplest and least prone to systematic errors possible; there is no need to measure the absolute flux, detector efficiency, etc. The flux transmitted through the sample T is related to the total cross section for the sample σ by the equation $T = e^{-n\sigma}$, where n is the sample thickness. The measurements on which the above calculation was based

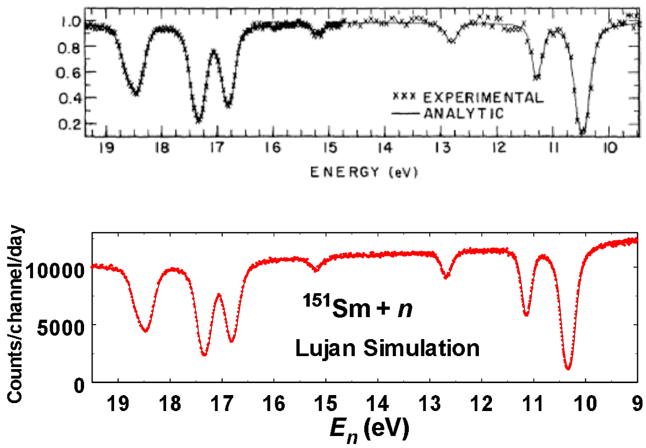

FIG. 2: Comparison of measured [1] transmission (top) to simulated sample-in counting rate (bottom) for a transmission experiment at the MLNSC using a 100-μg sample of ^{151}Sm. See text for details.

were made at a facility whose flux was orders of magnitude smaller than that available at the MLNSC. Scaling from the measured flux [8] at MLNSC, it should be possible to make measurements of similar quality on the same nuclide with a sample as small as 7 μg.

Representative simulation results for a 100-μg ^{151}Sm sample are compared to the data of Ref. [1] in Fig. 2. Simulations assumed a flight path length of 60 m, the same sample thickness used in Ref. [1], and the measured efficiency for a 1.27-cm thick GS20 ^{6}Li-glass-scintillator detector [9]. The same rates would be expected for a 10-μg sample in 10 days time. Statistical fluctuations in the counting rates were calculated by sampling from normal distributions with standard deviations equal to the square root of the average counting rate for each channel.

As can be seen from Fig. 2, the statistical precision predicted by the simulations often is much better than in the measurements of Ref. [1], especially considering that there often are fewer channels in the measured spectra.

There were approximately 10^8 counts/day, or 58 counts/pulse for E_n = 0.3–300 eV for this simulation, corresponding to an average of 130 μs between events.

The sample likely represents the most challenging aspect of the experiment. On the other hand, sample requirements for a transmission experiment are considerably more relaxed than those for capture or fission. For example, the sample can be loaded in a large shield at the fabrication facility and this container can be an integral part of the transmission apparatus. In addition, even liquid samples are possible (and sometimes may be preferred).

Because the main goal of the experiment is to measure parameters for as many resonances of the isotope of interest as possible, the sample should be as highly enriched as possible. This requirement can be circumvented to some extent by extra measurements for the contamination isotopes and/or measurements on the same sample at times spaced far enough apart that the isotopic composition of the sample has changed substantially due to radioactive decay. This technique was used, for example, to measure both the ^{249}Bk and ^{249}Cf total cross sections with a single sample [10].

Test experiments to measure actual counting rates and study backgrounds would be relatively easy to implement on, for example, flight path 5 at the MLNSC. Assuming a flight path length of 60 m, the proposed technique works best for nuclides having $D_0 \lesssim 20$ eV and half lives longer than a few days. Using D_0 values calculated with the default level-density model in TALYS [7], the following nuclides of high interest should be measurable: ^{88}Y, ^{107}Pd, ^{110m}Ag, ^{134}Cs, ^{147}Nd, ^{147}Pm, ^{145,146,147,148,149,150,152,154,155,156}Eu, ^{153}Gd, ^{160,161}Tb, ^{163,166}Ho, ^{169}Er, ^{167,168,170,171}Tm, ^{175}Yb, ^{179}Ta, ^{173,174}Lu, ^{185}W, ^{186}Re, ^{191}Os, ^{189,190,192}Ir, ^{193}Pt, and ^{195,196,198}Au.

The largest remaining uncertainty (in the NSM prediction of the capture cross section) is expected to be the p-wave neutron strength function S_1. However this parameter can likely be constrained to sufficient accuracy by systematics, or by measuring the average transmission in the unresolved region.

As many of the nuclides identified as being of high interest 20 or more years ago remain unmeasured, it should be possible to build a strong case for this new capability.

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